

# Optical probing of spin dynamics of two-dimensional and bulk electrons in a GaAs/AlGaAs heterojunction system

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We present time-resolved Kerr rotation measurements of electron spin dynamics in a GaAs/AlGaAs heterojunction system that contains a high-mobility two-dimensional electron gas (2DEG). Due to the complex layer structure of this material the Kerr rotation signals contain information from electron spins in three different layers: the 2DEG layer, a GaAs epilayer in the heterostructure, and the underlying GaAs substrate. The 2DEG electrons can be observed at low pump intensities, using that they have a less negative g-factor than electrons in bulk GaAs regions. At high pump intensities, the Kerr signals from the GaAs epilayer and the substrate can be distinguished when using a barrier between the two layers that blocks intermixing of the two electron populations. This allows for stronger pumping of the epilayer, which results in a shift of the effective g-factor. Thus, three populations can be distinguished using differences in g-factor. We support this interpretation by studying how the spin dynamics of each population has its unique dependence on temperature, and how they correlate with time-resolved reflectance signals.

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## I. INTRODUCTION

Spintronics is by now a well established field that aims to expand the potential of conventional electronics by exploiting the spin of electrons in addition to their charge<sup>1,2,3</sup>. The field attracted widespread attention with the proposal of a spin-based field effect transistor by Datta and Das<sup>4</sup> in 1990. Later, the discovery<sup>5</sup> of exceptionally long spin dephasing times in bulk *n*-GaAs prompted numerous investigations of spin dynamics in GaAs based systems. For double-sided quantum wells (QW) –not doped or with a two-dimensional electron gas (2DEG)– various studies with optical techniques aimed at understanding the more complex dynamics of 2D electron spin ensembles. The dependence of spin dephasing times and electron g-factors on parameters such as confinement energy, carrier density and crystal orientation has been investigated<sup>6,7,8,9,10,11,12,13</sup>.

Heterojunction systems, with a strongly asymmetric QW that contains a high-mobility 2DEG, also have long attracted considerable interest. These systems provide the highest values for electron mobility. Further, this material is of interest since the QW asymmetry results in a strong and tunable Rashba spin-orbit coupling, that can cancel Dresselhaus spin-orbit coupling<sup>14</sup>, and this gives access to control over spin relaxation and dephasing<sup>13,15,16,17,18,19,20,21,22</sup>. However, most of what is known about electron spin dynamics in these heterostructures was obtained using transport measurements<sup>14,23,24,25</sup>. Complementing this with magneto-optical pump-probe measurements brings the advantage that spin dynamics can be studied with ultra-high time resolution.

We report here on using time-resolved magneto-optical

Kerr rotation (TRKR)<sup>26</sup> to study the dynamics of spin ensembles in such a heterojunction QW system that contains a high-mobility 2DEG. Initial TRKR studies on such systems suggest that this may also provide an interesting platform for studying electron-electron interactions<sup>27,28</sup>. However, it is not yet well established in which regimes the Kerr response of 2DEG electrons can be reliably isolated from other contributions to the Kerr signal. The reason is that such multilayered GaAs/AlGaAs heterostructures have several GaAs layers that have an identical value for the gap. This results in optical readout signals that contain contributions from (photo-excited) electron populations in several layers.

For our heterostructure, three different layers can contribute to the observed Kerr signals from electron spins: the 2DEG layer, the bulk epitaxial *i*-GaAs layer that forms the heterojunction and the *i*-GaAs substrate. The response of these three layers occurs at nearly the same photon energy, hindering discrimination of layers by tuning the laser photon energy when using very short pulses. We exploit, instead, that one can use differences in the g-factor to isolate the signal from each population. The g-factor depends on the electron kinetic energy  $E$  (with respect to the bottom of the conduction band), which can be approximated as

$$g = g_0 + \gamma E, \quad (1)$$

where for bulk GaAs<sup>29,30</sup>  $g_0 \approx -0.44$  and  $\gamma \approx 6.3 \text{ eV}^{-1}$ . For 2DEG electrons these parameters were reported<sup>30</sup> as  $g_0 \approx -0.377$  and  $\gamma \approx 4.5 \text{ eV}^{-1}$ . Due to band filling by (photo)electrons, the g-factor thus shifts with an increase of the (quasi-)Fermi level as well. A related consequence is that the g-factors shifts towards less negative values with increasing the degree of confinement

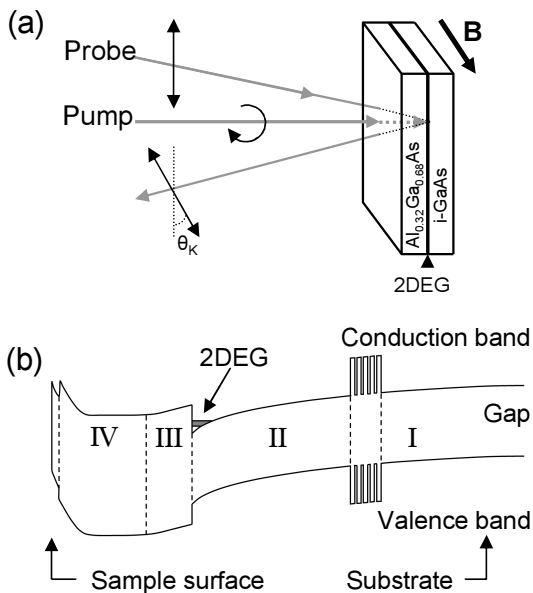


FIG. 1: (a) Schematic of the setup for time-resolved Kerr rotation measurements with pump and probe pulses incident on the 2DEG sample. The pump pulse is circularly polarized and the probe is linearly polarized. The probe pulses are incident on the sample at a small angle from the normal ( $\sim 2.3^\circ$ ). The applied magnetic field is oriented in the plane of the 2DEG. The rotation angle  $\theta_K$  of the linear probe polarization is measured as a function of the time delay between pump and probe pulses. (b) Schematic of the conduction and valence bands of the heterojunction system that contains a 2DEG (not to scale). The composition of the different layers I-IV is detailed in the main text. At the interface between layers II and III the conduction bands of the two materials bend, forming a potential well in layer II where the 2DEG is formed.

in a QW (possibly with an increased shift from wavefunctions penetrating AlGaAs barriers). However, it is not obvious that discrimination by g-factors can be applied without accounting for electron momentum relaxation and the intermixing of the populations in different layers<sup>31</sup> during the evolution of spin dynamics. Our measurement technique relies on using a pump pulse which inserts optically-oriented electron-spin populations well above the bottom of the conduction band, and subsequent momentum relaxation and diffusion of carriers can result in intractable Kerr signals. Nevertheless, our results show that one can define situations and regimes (defined by pump intensity) where the three spin populations can be studied without being hindered by these effects.

The g-factor of the 2DEG is well separated from that of the bulk layers, and our results show that the 2DEG population can be studied as an independent population when the density of photo-excited carriers does not exceed the 2DEG electron density from doping. The g-factor for the two bulk layers, on the other hand, is the same so a dif-

ferent approach is needed to distinguish the signal originating from each. Here we use that our heterostructure contains a barrier that blocks carrier diffusion between the two bulk *i*-GaAs layers. The effect of this is that the carrier density in the layer closest to the surface reaches a higher average value. The effective g-factor for the uppermost *i*-GaAs layer then acquires a less negative value (Eq. 1) than that of the underlying substrate. This can be applied in a regime where the density of photo-excited carriers exceeds the 2DEG electron density from doping, and under these conditions the signal from the 2DEG is suppressed by strong band filling in the 2DEG layer.

In Section II of this article we discuss the experimental methods and sample materials that we used. Results are presented and discussed in Section III. There, we first compare the Kerr response of our heterostructure to that of well-characterized<sup>5,32,33</sup> bulk *n*-GaAs material that is probed under identical conditions in our setup, and we highlight the differences. Next, we show that the response from the heterostructure can always be described as a superposition of signals from two independent electron populations, and we confirm that this is true for both the Kerr signals and for time-resolved reflectance signals that give insight in carrier dynamics of the populations. Measurements of spin dynamics are then analyzed in both the time and frequency domain, confirming that it is possible to treat the different electron populations as independent. In order to confirm that we correctly assign each of the observed g-factors to a particular population in the heterostructure we study how the g-factor, spin dephasing time, and carrier lifetime of each population depend on pump-photon density and temperature. Section IV presents a summary of the main findings and conclusions.

## II. METHODS

### A. Time-resolved probing of electron spins

In a semiconductor, a laser pulse can promote electrons from valence-band states to conduction-band states. In a III-V semiconductor like GaAs, such transitions obey well-defined selection rules. If the laser pulse is linearly polarized, the number of photo-excited electrons with their spin parallel to the beam's propagation direction equals the number of photo-excited electrons with their spin antiparallel to the beam's direction. If the laser pulse is circularly polarized, the number of photo-excited electrons with spin aligned along the beam's direction will exceed by 3 to 1 the number of electrons with spin aligned in the opposite direction (and vice versa for the opposite light helicity)<sup>34</sup>. A circularly polarized laser pulse can therefore generate a non-equilibrium population of spin polarized electrons in the conduction band of GaAs. This process is known as optical spin orientation.

In TRKR experiments, optical spin orientation with an ultrashort pump pulse is followed by optical spin probing

with a weaker laser pulse with linear polarization that is reflected on the sample. An unequal filling of the spin-up and spin-down conduction bands produced by the pump pulse gives rise to a transient difference in the absorption coefficient for right and left circularly polarized light (RCP and LCP respectively). Through Kramers-Kronig relation this also gives rise to a difference in the refractive index for RCP and LCP light. Such a difference in the refractive index gives a rotation of the linear probe polarization upon reflection on an interface of the sample, given that the linear polarization is a superposition of RCP and LCP light. This process is known as Kerr rotation, and the Kerr rotation angle  $\theta_K$  (for probing with a certain photon energy) is proportional to the expectation value for spin orientation parallel to the propagation direction of the incident probe beam. The Kerr rotation is strongest when the probe photon energy is near resonance with transitions to the states with unequal spin filling, but its magnitude and sign depend on the detuning with respect to exact resonance<sup>35,36</sup>, and interference effects and additional Faraday rotations in systems with reflections from multiple interfaces<sup>37</sup>. Note that TRKR signals thus predominantly reflect the properties of electrons at the quasi-Fermi level of the photo-induced electron population.

In addition to rotation, the reflected pulse may have obtained a certain degree of ellipticity, known in this case as Kerr ellipticity. In our experiments we study the reflected probe with a polarization bridge with balanced photodetectors<sup>26</sup>. The recorded signal is then proportional to the Kerr rotation, while Kerr ellipticity and reflectance changes give only third order corrections. Figure 1a depicts the configuration of the experimental setup with pump and probe beams incident on the sample, and the rotated linear polarization of a reflected probe pulse. A TRKR trace is obtained by plotting the Kerr rotation angle  $\theta_K$  of the reflected probe pulse as a function of the time delay between the pump and probe pulse (see for example Fig. 2).

In the presence of an in-plane external magnetic field (perpendicular to the initial orientation of the optically pumped spins), the injected spins precess around the field at the Larmor frequency  $\omega_L$ ,

$$\omega_L = g\mu_B B/\hbar, \quad (2)$$

where  $g$  is the electron g-factor,  $\mu_B$  is the Bohr magneton,  $B$  is the magnitude of the applied magnetic field, and  $\hbar$  is the reduced Planck constant. In TRKR traces this spin precession appears as oscillations of the Kerr angle at the Larmor frequency (Fig. 2b). Such traces can thus be used for determining the electron g-factor with Eq. 2.

We characterize the transient nature of TRKR traces with a Kerr signal decay time  $\tau_K$ . Note that this can be shorter than the spin dephasing time  $T_2^*$  for an electron ensemble. We restrict the use  $T_2^*$  for the case that the number of optically oriented electrons in the probe volume is constant. When this number is decreasing in time at a rate  $1/\tau_e$  due to processes as electron-hole re-

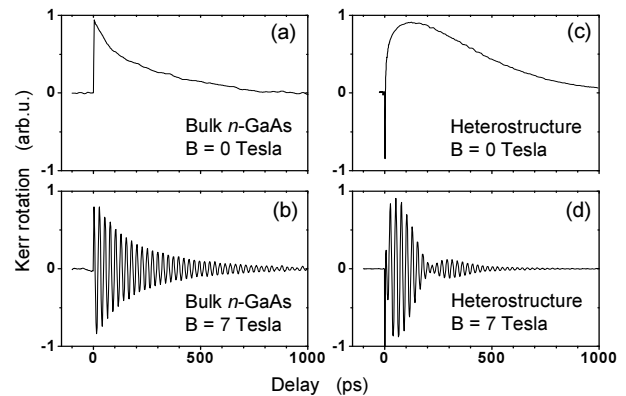


FIG. 2: Time-resolved Kerr rotation (TRKR) signal at 4.2 K from bulk  $n$ -GaAs at 0 Tesla (a) and 7 Tesla (b) and for the heterostructure containing a two-dimensional electron gas (2DEG), also at 0 Tesla (c) and 7 Tesla (d). The data shows considerable differences between the spin signals from the bulk and 2DEG sample. Most remarkable is the presence of a node (at  $\sim 200$  ps) in the Kerr oscillations measured at 7 Tesla on the 2DEG sample (plot d). Comparing plots (a) and (c) also clearly shows a slow increase of the Kerr signal for the 2DEG sample for delays in the range 0 to 100 ps (also present in the 7 Tesla data). Data taken with photon density of  $\sim 90 \cdot 10^{11}$  photons/cm<sup>2</sup> per pump pulse.

combination or diffusion out of the probe volume, the Kerr signal decays according to  $1/\tau_K = 1/T_2^* + 1/\tau_e$  (an exception to this rule applies to electron doped systems, where Kerr signals can live longer than the recombination time<sup>5</sup>). We obtain  $\tau_K$  from measuring TRKR traces at  $B = 0$  or from studying the envelope of oscillatory signals at  $B > 0$ . In such TRKR traces, hole spins only contribute to the signal at very short pump-probe delays (up to  $\sim 5$  ps), because ensembles of hole spins dephase very rapidly<sup>38,39,40,41</sup>. Thus, our TRKR measurements predominantly represent electron spin dynamics.

The dynamics of photo-excited carriers can be studied independently from their spin polarization with time-resolved reflectance ( $\Delta R$ ) measurement. For these measurements, a pump pulse with linearly polarized light is used. In this way, no net spin polarization is generated. Subsequently, a collinearly polarized probe pulse is incident on the sample. The pump-induced changes in sample reflectance are measured by comparing the intensity of the reflected probe pulse in the presence and absence of a preceding pump pulse. This method thus probes the presence of pump-induced electrons and holes at energies that are resonant with the probe photon energy, as a function of pump-probe delay. In time, photo-excited electrons and holes relax to the lowest available states, and electrons and holes recombine. This brings the sample reflectance back to its equilibrium value.

## B. Sample materials

Figure 1b depicts the profile of the valence band and conduction band of the heterostructure that we studied. Layer I is a (001) oriented *i*-GaAs substrate. On this substrate a multilayer buffer consisting of ten periods of alternating GaAs (5.2 nm) and AlAs (10.6 nm) layers was grown to smoothen the surface and to trap unintentional impurities. Layer II, the accumulation layer, consist of 933.0 nm of undoped GaAs. A spacer layer (III) is formed by 36.8 nm of undoped  $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$  grown on top of the accumulation layer. The donor layer (IV) consists of 71.9 nm of Si-doped  $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$  with  $\sim 1 \cdot 10^{18}$  dopants/ $\text{cm}^3$ . The heterostructure is capped with 5.5 nm of *n*-GaAs. The two AlGaAs layers have a bandgap that is larger than all the photon energies that we used in our experiments. Due to quantum confinement the  $\sim 5$  nm GaAs layers of the multilayer buffer and capping layer also only have optical transitions at energies above the photon energies that we use.

In this material a 2DEG is formed since dopant electrons from the donor layer (IV) reduce their energy by relaxing into the conduction band of the narrow bandgap accumulation layer (II). These excess carriers are held at the interface by electrostatic attraction from the ionized donors. The conduction band of the accumulation layer is pulled below the Fermi level only for a  $\sim 15$  nm wide layer near the interface, which results in the 2DEG at this position (estimate obtained from solving the coupled 1D Poisson-Schrödinger equations for the heterostructure<sup>42</sup>). Electron transport experiments on the 2DEG at 4.2 K, after illumination, gave  $2.7 \cdot 10^6$   $\text{cm}^2/\text{Vs}$  for electron mobility, and  $4.7 \cdot 10^{11}$   $\text{cm}^{-2}$  for the electron density. We obtained nearly identical values with optical studies on our 2DEG material<sup>43</sup>.

The bulk *n*-GaAs reference material we used had a doping concentration of  $(2.4 \pm 0.2) \cdot 10^{16}$   $\text{cm}^{-3}$ . Several experiments on this material have shown that at this doping concentration very long spin dephasing times can be obtained with Kerr studies, and the dependence of spin dephasing times on various experimental conditions has been characterized<sup>5,32,33</sup>.

## C. Experimental setup

The measurement setup comprises a magneto-optical flow cryostat, pulsed laser, polarization optics, and detection system. Sample temperatures are varied between 4.2 K and 100 K in the flow cryostat. The cryostat is equipped with a superconducting magnet used to apply fields up to 7 Tesla. The magnetic field is set parallel to the plane of the sample while the laser pulses are incident normal to the sample plane as shown in Fig. 1a. We used two different pulsed laser systems. Most of the presented results were obtained with a cavity-dumped mode-locked Ti:sapphire laser (further named Laser 1) with 15 fs pulses corresponding to a fixed spectrum extending from

740 nm to 880 nm, (repetition rates ranging from 4 MHz to 80 MHz). This allows for wavelength tuning with 10 nm bandpass interference filters. The central wavelength for both pump and probe beam can thus be chosen independently. After filtering, the pulse spectrums have a significant amplitude in a 19 meV window. The filters are followed by prism compressors to ensure pulse durations with a full width at half maximum of approximately 120 fs at the sample. Unless stated otherwise, we present measurements from using Laser 1 with the pump and probe pulse wavelengths centered at 780 nm and 820 nm respectively. We reproduced most results in a setup with a tunable Ti:sapphire laser (further named Laser 2) with  $\sim 150$  fs pulses at 80 MHz repetition rate. The spectrum of pulses is wider than Fourier-transform limited, with significant amplitudes over a  $\sim 15$  meV window. With this laser pump and probe pulses were always centered at the same wavelength.

In each case, two beams derived from the laser are used as pump and probe with power ratio 24:1 or 4:1. We checked that all our results were at intensities below the regime where saturation effects occur: Kerr signals were always proportional to both the pump and probe intensity, and the probe was always non-invasive in the sense that the observed decay times did not depend on probe intensity. We report the intensity of pump pulses in units of photons/ $\text{cm}^2$  per pulse, because it has relevance with respect to the two-dimensional electron density from doping. Note that this is the number of photons that is incident on the sample surface, and that the number is lower inside the sample due to reflection on each interface of the heterostructure. Glan-Thompson polarizers on both pump and probe beam lines are used to produce well defined polarizations. The pump-probe delay is varied using a retro-reflector mounted on a stepper-motor driven translation stage. The two beams are focused on the sample with a 25 cm focal length spherical mirror which gives spots with a diameter of approximately 150  $\mu\text{m}$  full-width at base level.

In order to measure the transient Kerr rotation of the sample, a photo-elastic modulator (PEM) is used to modulate the polarization of the pump beam between RCP and LCP at a rate of 50 kHz. Besides improvement of signal-to-noise, this is crucial for avoiding dynamical nuclear polarization effects<sup>33</sup>, and it rules out phase offsets in Kerr signals from interference effects in case of reflections from multiple interfaces<sup>37</sup>. The probe beam remains linearly polarized. The reflected probe is analyzed with a polarization bridge, which decomposes the probe beam into two orthogonal polarizations with a half-wave plate and a Wollaston prism, and which detects both these components with a pair of balanced photodetectors. This bridge is tuned to give a zero difference signal from the two detectors for probe pulses with zero Kerr rotation, and this difference signal is recorded with lock-in detection at the PEM frequency. For measurements with overlapping pump and pulse spectrums, we employed a double modulation technique by adding an optical chop-

per in the probe beam (at  $f_C \sim 1$  kHz) and recording a side band at  $f_C$  from the 50 kHz PEM frequency. In both cases, the signal is proportional to the Kerr rotation angle  $\theta_K$ .

With this setup it is possible to measure transient reflectance ( $\Delta R$ ) and TRKR under identical conditions. For  $\Delta R$  measurements the pump and probe beams have parallel linear polarizations. The reflected probe beam is now sent directly to a single photodetector, while the pump beam is modulated using an optical chopper. Modulations of the reflected probe intensity at the chopping frequency are then recorded with lock-in techniques. This gives a signal that is proportional to pump induced reflectance change of the sample.

#### D. Relaxation processes and time scales

Before discussing the experimental data it is useful to discuss a hierarchy of timescales that is relevant for interpreting our results. The electron spin dynamics occurs at time scales of tens to hundreds of picoseconds. For example, the Larmor precession period of electrons is of the order of 25 ps at 7 Tesla, and we observe electron spin dephasing times for the 2DEG in the range of 100 ps. The duration of the pump and probe pulses (approximately 120 fs) therefore provide sufficient time resolution to resolve all important spin dynamics. Hole spins dephase within  $\sim 5$  picoseconds<sup>38,39,40,41</sup>. Thus, the Kerr signal after a few picoseconds contains information about electron spins only. Most of the data shown uses delay steps of  $\sim 1$  ps with delays up to 1 ns, which allows us to capture all relevant dynamics of electron spins.

The timescales of other electron and hole relaxation processes are very diverse, ranging from a few tens of femtoseconds to nanoseconds. We typically use a pump pulse with the photon energy centered at 780 nm, pumping electrons in GaAs layers at about 63 meV above the bottom of the conduction band. This leaves a non-equilibrium and non-thermal carrier distribution in the conduction and valence bands of the GaAs layers. Within the first hundreds of femtoseconds, conduction band electrons (and valence band holes) thermalize into a Fermi-Dirac distribution with a carrier temperature that is much higher than the lattice temperature. Subsequently, electrons cool towards the lattice temperature. The initial cooling of electrons<sup>44,45,46,47,48,49</sup>, with excess kinetic energy of more than 10 meV occurs very rapidly due to rapid momentum relaxation (within 10 ps). Once the excess kinetic energies drop below  $\sim 10$  meV, further electron cooling slows down to a decay time of approximately 100 ps depending on electron density. Note however, that we use probe pulses with about 19 meV spectral width, with the probe spectrum centered at 820 nm, just below the bottom of the conduction band of GaAs. Consequently, most electrons relax into the spectral window of the probe within 10 ps, and the final cooling stage in the time interval from 10 to 100 ps has little influence on

our Kerr and reflectance signals. Thus, the fast carrier thermalization and initial cooling allows for pumping the sample with high-energy photons (convenient for blocking of pump light that scatters into the probe channel by spectral filtering, and it ensures that the penetration depth of pump light for the top GaAs layers is less than  $1 \mu\text{m}$ ). Right after a pump pulse, there will be a steep gradient in the photo-electron density along the direction of the pump over the length of the penetration depth. This equilibrates by electron diffusion along the beam direction. In the accumulation layer this takes place within the first 50 ps for high pump intensities, but may occur much slower<sup>37</sup> for low pump intensities. For our spot sizes, electron diffusion into directions perpendicular to the laser beam propagation does not significantly change the electron density profile.

Other carrier processes that take place in timescales between 1 ps and 1 ns are exciton formation and carrier recombination. Within the first hundred picoseconds electrons and holes form excitons. This implies a further reduction of the initial energy of the photo-excited electrons by a few meV, but this occurs within the spectral window of our probe, and these processes do therefore not strongly influence our  $\Delta R$  and TRKR traces. Electron-hole recombination, on the other hand, is directly visible in  $\Delta R$  measurements. Obviously, recombination also results in loss of Kerr signal, especially in undoped samples or under conditions where the photo-excited electron density exceeds the electron density due to doping<sup>26</sup>. With time- and spectrally-resolved photoluminescence, we measured that the exciton recombination time for our 2DEG sample at 4.2 K is greater than 2 ns in the accumulation layer, while it is approximately 360 ps for the substrate<sup>43</sup>. Both of these timescales become shorter with increasing pump intensity. As a function of temperature the 2DEG recombination time increases, while it decreases for bulk GaAs layers and  $n$ -GaAs.

### III. RESULTS AND DISCUSSION

#### A. Kerr signals from a GaAs/AlGaAs heterojunction system

In order to highlight the Kerr response that is characteristic for the heterostructure, we compare it to Kerr measurements on the bulk  $n$ -GaAs sample that were obtained under identical conditions. Figure 2 shows in the left column TRKR signals from the bulk  $n$ -GaAs sample. Its Kerr signal at 0 Tesla closely resembles a mono-exponential decay. The signal at 7 Tesla shows oscillations at the Larmor precession frequency for electrons in GaAs ( $g$ -factor is  $|g| \approx 0.44$ <sup>50</sup>), with an envelope that shows again mono-exponential decay<sup>51</sup>. The transient TRKR signals from the heterostructure, on the other hand, are more complex. At 7 Tesla (Fig. 2d), the Kerr response shows a node (at  $\sim 200$  ps), indicating a beating between different precession frequencies. Addi-

tionally, the envelope shows a slow increase in the delay range from 0 to 100 ps, which is also observed at 0 Tesla (Fig. 2c). The *in-situ* comparison with bulk *n*-GaAs material is important for concluding that the observed beatings in TRKR traces are characteristic for the heterostructure, given that beatings can also appear in Kerr oscillations from *n*-GaAs under certain measurement conditions<sup>33</sup>.

The heterostructure data in Figs. 2c,d also show a very sharp spike at very short delays, with a decay time of about 3 ps. It is also present in several other TRKR traces from the heterostructure that we will present and it was also observed with the *n*-GaAs sample at weaker pump intensities. It is longer than the duration of pump-probe overlap, and therefore not an optically induced Stark effect<sup>36</sup>. Instead, it occurs at a timescale that is consistent with hole spin dephasing, and hot-electron momentum relaxation. This part of the Kerr signal is therefore possibly influenced by these effects. We further focus on the Kerr signals at delays of 5 ps and longer, and do not include this spike in further analysis.

Figures 3a-c show that the beatings and slow onset in the heterostructure Kerr signals appear for a wide range of pump intensities. However, the exact appearance clearly depends on pump intensity. In what follows we will show that all three traces in Figs. 3a-c can be described as the superposition of two oscillatory signals with mono-exponential decay remarkably well. Each of the two contributions then results from a different electron population, each with its own decay time and effective g-factor. At the highest pump intensities (Fig. 3a), the signal is dominated by two bulk *i*-GaAs electron populations, one in the substrate (layer I in Fig. 1b) and one in the accumulation layer (II). At the lowest pump intensity, (Fig. 3c) the signal is dominated by a population in the 2DEG quantum well, and a second one with bulk *i*-GaAs characteristics.

Figure 3g shows the Fourier transform of the trace in Fig. 3a. The amplitude spectrum shows two distinct peaks. It can be fit very well with two Lorentzians, and the same holds for the Fourier transform of the traces in Figs. 3b,c. This suggests that one can fit the traces in Fig. 3a-c in the time-domain with a superposition of two mono-exponentially decaying cosine functions. That also builds on observations in *n*-GaAs where TRKR signals from single electron population with a single g-factor (see also Fig. 2b) can be described by a mono-exponentially decaying cosine function<sup>5</sup>. For this fitting we use

$$\theta_K = A_1 \exp(-t/\tau_{K1}) \cos(2\pi\omega_{L1}t + \phi_1) + A_2 \exp(-t/\tau_{K2}) \cos(2\pi\omega_{L2}t + \phi_2). \quad (3)$$

Here  $t$  is the pump-probe delay,  $\tau_{K1}$  ( $\tau_{K2}$ ) and  $A_1$  ( $A_2$ ) are, respectively, the Kerr signal decay time and the Kerr rotation amplitude for each population. Similarly,  $\omega_{L1}$  and  $\omega_{L2}$  are the Larmor frequencies and  $\phi_1$  and  $\phi_2$  are apparent initial phases<sup>52</sup> for spin precession. For fitting we only use data for  $t > 5$  ps.

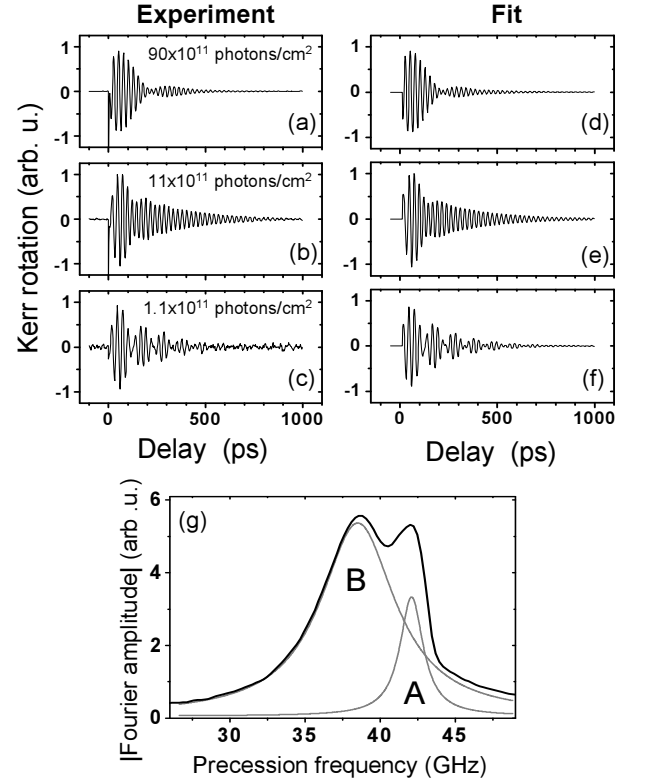


FIG. 3: Time-resolved Kerr rotation signal of the 2DEG sample for various pump photon densities. The left column, plots (a)-(c), show the sample response at photon densities per pump pulse as labeled. The pump-photon density strongly influences the beating pattern in the Kerr oscillations. The right column, plots (d)-(f), show fits to the data in (a)-(c) using Eq. 3 (see main text for details). Plot (g) shows a Fourier transform (black line) of the TRKR data of plot (a), showing two distinct precession frequencies (with labels for populations A and B as in Fig. 5). Good fits to the spectrum can be obtained using a superposition of two Lorentzians (gray lines). Data taken at 7 Tesla and 4.2 K.

The fitting results of applying this to the experimental traces of Fig. 3a-c are presented in Fig. 3d-f. The excellent fits demonstrate that the total signal can be indeed described as being composed of two distinct contributions from two different populations. We find that we can obtain very good fits assuming two populations only for all pump intensities. A similar approach that assumes three populations does not improve the fits significantly. This is a remarkable result, given that the Kerr response of our heterostructure does not necessarily split into two distinct contributions. The continuous character of the accumulation layer (II) from quantum well to bulk epilayer, followed by a bulk substrate, could give more complex signals with signatures of relaxation processes and mixing effects between populations<sup>31</sup>. We find that the sign of the amplitudes  $A_1$  and  $A_2$  of the two contributions is always opposite (for  $\phi_1$  and  $\phi_2$  close to zero) which corresponds to Kerr rotations of opposite

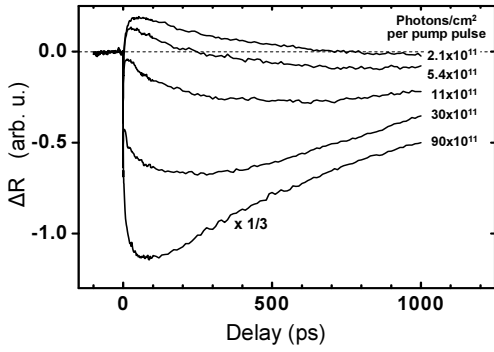


FIG. 4: Transient reflectance ( $\Delta R$ ) traces from the 2DEG sample at different pump-photon densities. Data taken at 4.2 K and 0 Tesla.

sign at  $t = 0$ . Consequently, the slow signal increase at early delays and the beatings in our Kerr oscillations have a common origin. The sign of the Kerr response depends on detuning from resonance, but we predominantly probe slightly red-detuned for all populations. The opposite sign is therefore most likely from interference effects that play a role for the sign of Kerr response with reflections from multiple interfaces<sup>37</sup>.

Further evidence that these excellent fits in the time domain indeed reflect that the physics that underlies our Kerr signals is that of two independent populations, each with a different but (nearly<sup>50</sup>) constant g-factor, comes from Fourier analysis of the traces. Scenario's that include significant spreads or strongly drifting g-factors due to momentum relaxation can be ruled out based on the phase spectrum of Fourier transformed TRKR signals. In addition, direct measurements of the momentum-relaxation dynamics support this interpretation.

We applied time-resolved reflectance measurements for these studies of the momentum-relaxation dynamics. It provides a direct measure for the momentum relaxation of electron populations into the spectral window of the probe and subsequent electron-hole recombination. Figure 4 shows  $\Delta R$  as a function of pump-probe delay measured on the heterostructure sample using different pump intensities. At high pump-photon densities,  $\Delta R$  first reaches a negative peak before decaying back to zero. At low pump-photon densities, the signal first shows a positive  $\Delta R$  but at later delays  $\Delta R$  obtains a negative value with only very slow decay back to zero.

Also here we find that we can describe the full set of  $\Delta R$  traces for  $t > 10$  ps as the sum of two mono-exponentially decaying responses of opposite sign: one response with positive  $\Delta R$  that is short lived and another response with negative  $\Delta R$  that is long lived. In addition, there is for each trace during  $0 < t \lesssim 10$  ps an onset proportional to  $1 - \exp(-t/\tau_{mr})$  with a decay  $\tau_{mr} \lesssim 10$  ps. This is consistent with the initial momentum relaxation of pumped electrons into the spectral

window of the probe. For  $t > 10$  ps, we obtain excellent fits with an approach analogous to fitting TRKR signals with Eq. 3 (but with the cosine factors set to 1). The difference in shape for traces recorded at different pump intensities is then predominantly due a shift in the relative weight of the two contributions.

At the lowest pump intensities, the signal is first dominated by a short lived ( $\sim 360$  ps) positive contribution to  $\Delta R$ . At later delays a longer lived (2-3 ns) negative contribution dominates the signal. This is consistent with the photoluminescence results<sup>43</sup> if we assign the positive short-lived contribution to the substrate, and the negative long-lived contribution to the 2DEG layer. Going to higher pump intensities, the relative weight of the negative contribution (ratio  $|A_2/A_1|$ , with  $A_2$  the amplitude of the negative contribution) increases from  $\sim 0.5$  to  $\sim 5$ . This is consistent with an increased importance for the accumulation layer (II) that can be expected. In addition, the decay time for negative long-lived population decreases to  $\sim 1$  ns. Note, however, that at high pump intensity there is significant band-filling in the accumulation layer (II), so it should now be considered the recombination time for an electron population that is extended throughout this layer instead of a 2DEG population. At the high pump intensities, the onset of the strong negative contribution (with onset proportional to  $1 - \exp(-t/\tau_{mr})$ ) curves over into the weak positive contribution with a decay time of  $\lesssim 360$  ps, which makes it impossible to reliably fit this latter time scale for high-pump intensities. When increasing the temperature to 100 K, the life time of the negative contribution gradually shortens to  $\sim 100$  ps, while the life time of the positive contribution grows to  $\sim 8$  ns.

Thus, we find that both TRKR signals and  $\Delta R$  signals can be described very accurately as the superimposed response of two different electron populations. Moreover, the relative amplitudes  $A_1$  and  $A_2$  of populations that are observed with TRKR correlate with those observed in  $\Delta R$  signals (further discussed at Fig. 5 below). The fact that the signal or signal-envelope for each of the two contributions is well described by mono-exponential decay confirms that photo-excited electrons cool within about 10 ps to a temperature where all electrons are at energies within the spectral window of the probe. Further cooling only causes small changes in the average value and spread of g-factors and are masked by the relatively short decay times of the Kerr signals. Further, we observe that for the substrate contribution to the signals, the decay times for the reflectance signals and Kerr signals are comparable. This means that the loss of Kerr signals is here for a significant part due to electron-hole recombination.

Although the TRKR traces from the heterostructure can always be fit assuming two spin populations, a total of three different spin populations show up in the TRKR measurements when these are studied for a range of pump pulse intensities. From such data, we analyze the precession frequencies of the different spin populations, both with Fourier analysis as in Fig. 3g, and fitting in the

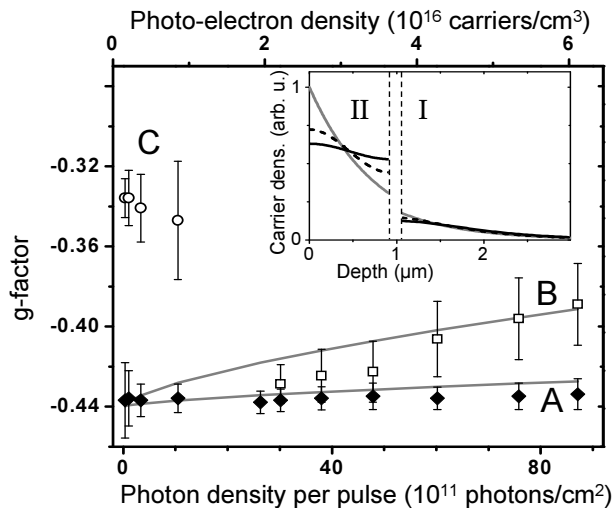


FIG. 5: The g-factors of spin populations observed in the 2DEG sample as a function of pump-pulse photon density (bottom axis) and the estimated photo-excited electron density in the accumulation layer (top axis). At high photon densities two spin precession modes can be resolved with g-factors  $|g| \approx 0.44$  (solid diamonds, population A) and  $|g| \approx 0.39$  (open squares, population B). We ascribe these g-factors to electrons in the *i*-GaAs substrate (layer I) and electrons in the accumulation layer (layer II) respectively. The gray lines through the data points of A and B are calculated g-factor values (see main text for details). A third precession mode (open circles, population C) is observed at the lowest pump-photon densities and corresponds to electrons in the 2DEG. Data obtained under conditions as in Fig. 3. The inset shows the calculated density of photo-excited electrons in the accumulation layer (layer II) and in the substrate (layer I) as a function of depth from the top of the accumulation layer. The dashed vertical lines at  $0.933 \mu\text{m}$  show the position of the multilayer buffer. The gray line shows the calculated electron density immediately after photo excitation with a 780 nm pump pulse. The dashed black line (solid black line) shows the electron density 20 ps (50 ps) after photo excitation.

time domain (Eq. 3). Figure 5 shows which g-factors are observed as a function of pump-pulse intensity. Two spin populations show up at high pump-photon density (labeled A and B) a third population is only observed at low pump-photon density (labeled C). The ratio of contributions  $|A_C/A_A|$ , going over into  $|A_B/A_A|$ , increases from  $\lesssim 1$  to  $\sim 5$  with increasing pump intensity. It thus closely follows the ratio  $|A_2/A_1|$  discussed for Fig. 4. This provides the first evidence that signal contributions B and C come from the accumulation layer, and A from the substrate. In the next Sections (III B and III C) we further discuss the origin of these three spin populations. We also analyze the physics that results in the fact that these populations behave independently, and the spin dynamics of each population. The error bars indicate here the uncertainty in position for the two Lorentzian peak shapes that we fit on the spectrums, and thus also give an indication for the Kerr decay times for each contribu-

tion (the smallest error bars corresponding to  $\sim 250$  ps, the largest to  $\sim 70$  ps).

## B. Kerr signals from *i*-GaAs populations

The value of the g-factor of each population gives a strong indication of the medium that the electrons populate. The g-factors of the different semiconductor materials that make up the heterostructure are well known. The g-factor of electrons in bulk GaAs is approximately  $-0.44^{53}$ , and electrons in heterojunction 2DEGs and 15 to 20 nm wide GaAs QWs have a g-factor of about  $-0.36^{11,54,55,56,57}$ . For electrons in the 5.5 nm wide GaAs layers  $g \approx +0.15$ , and for<sup>58</sup> bulk  $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$  layers  $g \approx +0.5^{53}$  (but electron populations in these latter two layers are not interacting with the photon energies that we use).

The value of the g-factor of population A is  $|g| \approx 0.44$  at all pump-photon densities. We therefore associate population A with electrons in bulk GaAs. Measurements of the temperature dependence of the g-factor of population A, discussed below, also indicate that these electrons populate a bulk layer of GaAs. Time-resolved Kerr rotation measurements cannot directly determine the sign of g-factors. However, the g-factor of GaAs is known to be negative<sup>53</sup> so we assign a negative value to the g-factor of population A.

The g-factor of population B shows a strong dependence on pump-photon density. At the highest pump-photon densities the g-factor of population B is  $|g| \approx 0.39$  (while the signal contribution B is here about 5 times stronger than A with its Kerr signal decay time as short as  $\sim 70$  ps, see also the broad peak in Fig. 3g). As the photon density is reduced, the g-factor of population B approaches the value of  $|g| \approx 0.44$ . This shows that this population also corresponds to electrons in bulk GaAs, and we again assign a negative value to its g-factor. There are two bulk *i*-GaAs layers in the heterostructure: the accumulation layer (II, see Fig. 1) and the substrate (I). The analysis in the next paragraphs demonstrates that population A corresponds to electrons in layer I, and that population B corresponds to electrons in layer II. Population C shows up in the TRKR measurements only at low pump-photon densities, and corresponds to 2DEG electrons, as will be further discussed in Section III C.

At high pump-photon densities, the response from the two *i*-GaAs layers can be distinguished because different average electron densities are excited in each layer by the pump pulse, which results in different g-factors (Eq. 1). In the following discussion we will show how different average electron densities are obtained thanks to a pump beam with short penetration depth and the multilayer buffer between layer I and layer II. At 4.2 K the absorption coefficient of 780 nm light in GaAs gives a penetration depth of approximately  $0.77 \mu\text{m}^{59}$ . The density of photo-excited electrons that is present immediately after a pump pulse decays exponentially as a function of



depth into the GaAs layers over this length scale. Thus, most of the incident pump photons are absorbed in the accumulation layer (II), giving a much higher concentration of photo-excited electrons in this layer than in the substrate (I). The gray line in the inset in Fig. 5 shows the density of photo-excited electrons as a function of depth immediately after the absorption of a pump pulse. This strong gradient in the photo-electron density will rapidly equilibrate due to diffusion. The inset also shows the calculated electron density as a function of depth 20 ps and 50 ps later. We used here a bulk diffusion constant<sup>43,60</sup> of  $30 \text{ cm}^2/\text{s}$ , but the results of the model that we present below are quite independent from the exact value of diffusion constant that is used. In this calculation, the influence of the space-charge potential in the accumulation layer is ignored. This is justified at high pump-photon densities because the space-charge potential is screened by the photo-excited carriers. In addition, carrier-recombination effects are ignored since these occur at timescales longer than the considered diffusion times. Note that the penetration depth for part of the probe spectrum (centered at 820 nm) is considerably longer than that of the pump, such that the Kerr response of the deepest photo-excited electrons is not strongly attenuated with respect to electrons near the surface. In addition, the reflection on the multilayer buffer layer ensures that there is an enhanced Kerr response for electrons in its direct vicinity.

The dashed vertical line in the inset in Fig. 5 at  $0.933 \mu\text{m}$  represents the position of the multilayer buffer, which acts as a barrier that blocks electron diffusion. The calculated electron density profiles at 20 ps and 50 ps show that the electron density, and thus the quasi-Fermi level in the two layers, becomes discontinuous at the multilayer buffer. The accumulation layer reaches a much higher average electron density than the substrate (see also top axis of Fig. 5 for estimated values), and thereby a much higher value for the of the quasi-Fermi level that is established after tens of picoseconds when most electrons are in the lowest available conduction band states (besides spin relaxation) due to cooling and diffusion. The average electron g-factor of a population that is observed in TRKR signals depends on the quasi-Fermi level as in Eq. 1. We used this to calculate the expected g-factors of electrons near the quasi-Fermi level for the population in the accumulation layer (II) and the substrate (I), without any adjustable parameters<sup>38</sup>. When calculating a photo-electron density from pump-photon density that is incident on the heterostructure surface we accounted for an estimate of the reflection on each interface in the heterostructure. These calculated g-factors are also plotted in Fig. 5. The good agreement between the measured and calculated g-factors for populations A and B confirms that the two populations are indeed electrons in the accumulation layer and the substrate.

The above discussion shows that the ability to distinguish the two populations relies on the short penetration depth for the pump pulse. Our results did not change sig-

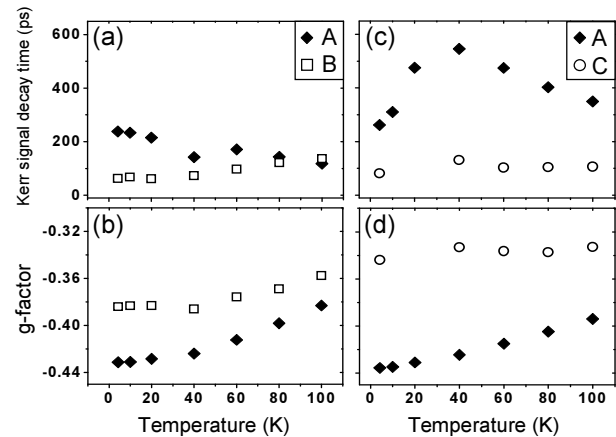


FIG. 6: Kerr signal decay times (top row) and g-factors (bottom row) as a function of temperature, measured at pump photon densities of  $\sim 100 \cdot 10^{11} \text{ photons/cm}^2$  (left column) and  $\sim 3.6 \cdot 10^{11} \text{ photons/cm}^2$  (right column) in the heterostructure sample. Electron populations A, B, and C (see also Fig. 5) are indicated by open squares, solid diamonds, and open circles respectively. The data points were obtained from fits with Eq. 3 to TRKR measurements taken at 7 Tesla.

nificantly when using 800 nm pump and 820 nm probe photons (also showing that the initial rapid momentum relaxation does not result in significant spin dephasing). However, when using 820 nm photons for both pump and probe with Laser 1, we did no longer observe the beatings in our TRKR oscillations. Under these conditions, the relative contribution from the population in the substrate is much higher due to deeper penetration of pump light into GaAs (the penetration depth increases beyond  $3 \mu\text{m}$  when the wavelength increases beyond  $820 \text{ nm}$ <sup>59</sup>). This prohibits an analysis of the contribution to the Kerr signal from the top GaAs layers of the heterostructure. We could, nevertheless, fully reproduce our results in a mono-color experiment, explored with the tunable Laser 2. Also here, we had to ensure that the full spectrum of laser pulses had a short penetration depth. We could get results with a strong contribution from the accumulation layer to the Kerr response (both at high and low pump intensities) by setting the central laser wavelength at least  $\sim 20 \text{ meV}$  above the bottom of the conduction band. Momentum relaxation brings electrons again rapidly near the bottom of the conduction band, and probing then occurs predominantly with the low-energy wing of our pulse spectrum. When tuning the central wavelength above  $\sim 810 \text{ nm}$ , the Kerr response was again fully dominated by electrons in the substrate.

As an additional check for our interpretation in this regime with high pump-photon densities we studied how the Kerr signals depend on temperature  $T$  (presented in Fig. 6) and magnetic field  $B$  (only discussed). The Kerr signal decay times for populations A and B as a function of temperature are presented in Fig. 6a, and the corresponding g-factors in Fig. 6b. These results were ob-

tained from fitting Eq. 3 to TRKR measurements taken at  $\sim 100 \cdot 10^{11}$  photons/cm<sup>2</sup> per pump pulse. The temperature dependence of the g-factors of populations A and B is in agreement with observations by Oestreich *et al.* on GaAs bulk samples<sup>38</sup>, confirming that populations A and B correspond to electrons with bulk GaAs characteristics. The g-factor of population B increases with temperature in a manner similar to population A. However, with increasing temperature the difference decreases. This is consistent with a broadening of the quasi-Fermi level with temperature, which increases the average electron kinetic energy  $E$  and (by Eq. 1) brings the g-factor values closer to zero. At the highest temperatures, the broadening of the quasi-Fermi level starts to become larger than the difference in quasi-Fermi level for populations A and B, which results in a smaller difference between the two g-factor values.

The Kerr signal decay times  $\tau_{KA}$  and  $\tau_{KB}$  of populations A and B show opposite trends as a function of temperature. At 4.2 K the decay time of population A is approximately 240 ps. As the temperature is raised this value decreases monotonically down to  $\sim 120$  ps. The signal decay time of population B, on the other hand, increases from  $\sim 70$  ps at 4.2 K to  $\sim 140$  ps at 100 K. We analyzed that several mechanisms contribute to these Kerr signal decay times, and full understanding goes beyond the scope of this article. Instead, we will only discuss a few trends and typical values. The following three mechanisms contribute to the Kerr signal decay rates that we observe:

- i) Electron-hole recombination. This directly contributes to loss of Kerr signals at the electron-hole recombination rate.
- ii) Precessional dephasing due to a spread  $\Delta g$  in g-factor values. At high pump-photon densities, band filling gives that the spin-oriented electrons have a spread in electron kinetic energy  $E$ , which (by Eq. 1) directly results in a spread  $\Delta g$ . This results in a dephasing rate<sup>5</sup>  $1/T_2^* = \Delta g \mu_B B / \sqrt{2} \hbar$ .
- iii) The D'Yakonov-Perel' (DP) spin dephasing mechanism<sup>63,64</sup>, as a result of the cubic Dresselhaus spin-orbit coupling in GaAs: The spins of electrons in motion experience a  $k$ -vector dependent effective magnetic field. Consequently, random momentum scattering (at a rate  $1/\tau_p$ ) randomizes spin states by precession. At low temperatures and high electron densities, this effect contributes to spin dephasing at a rate (for a review see Ref. 2)  $1/T_2^* \propto \tau_p \gamma_{cub}^2 n_{3D}^2$ , where  $\gamma_{cub}$  is the cubic Dresselhaus constant, and  $n_{3D}$  the bulk (photo-)electron density. The rate of this effect typically decreases with increasing magnetic field if  $\tau_p$  is long enough to yield cyclotron motion. We estimate the DP rates with a numerical Monte-Carlo approach, that we described in detail in Ref. 65.

In our experiments, these contributions are often of similar magnitude, but the weights shift with temperature, field and electron density. At 4.2 K and 7 T, and for pumping at  $\sim 100 \cdot 10^{11}$  photons/cm<sup>2</sup>, the estimate

for the precessional dephasing rate by  $\Delta g$  gives about  $(1 \text{ ns})^{-1}$  for population A, and  $(150 \text{ ps})^{-1}$  for population B, thus not fully explaining the observed decay rates. In addition, both  $\tau_{KA}$  and  $\tau_{KB}$  gradually increase by a factor  $\sim 3$  when lowering  $B$  towards zero. This dependence is weaker than  $1/B$  for both  $\tau_{KA}$  and  $\tau_{KB}$ , confirming that a spread in  $\Delta g$  is not fully responsible for the observed decay. The electron-hole recombination time for population A is about  $\lesssim 300$  ps at 4.2 K (Section III A), and was observed to decrease to about 100 ps at 100 K. It thereby provides the dominant rate contributing to the decay of A, and also explains the temperature dependence of  $\tau_{KA}$  in Fig. 6a. This is supported by the fact that the  $n$ -GaAs reference sample (measured under identical conditions) shows here very similar decay times and temperature dependence as population A.

As said, the dependence of  $\tau_{KB}$  on temperature is opposite to that of  $\tau_{KA}$ . For population B (in the accumulation layer), the electron-hole recombination time is much longer than for A (Section III A), and increasing from  $\sim 1$  ns to  $\sim 8$  ns when increasing  $T$  to 100 K. Thus, electron-hole recombination barely contributes to the observed  $\tau_{KB}$  values. However, due to the much higher electron density for B, the DP mechanism now probably contributes significantly. The Monte-Carlo simulation (for 0 K, 7 T,  $n_{3D} = 7 \cdot 10^{16} \text{ cm}^{-3}$ , and the typical value<sup>14</sup>  $\gamma_{cub} = 30 \cdot 10^{16} \text{ eV nm}^3$ ) give a DP rate of  $(200 \text{ ps})^{-1}$  if we assume a high mobility of  $4 \cdot 10^5 \text{ cm}^2/\text{Vs}$  (giving  $\tau_p = 150 \text{ ps}$ ). Such high mobility values were indeed directly observed in transient grating experiments on our sample material<sup>43</sup> at these high photon-electron densities, and due to the fact that population B is in a pure undoped layer of epitaxial quality. We can also explain the temperature dependence of B in Fig. 6a when making this assumption. The observed momentum scattering rate  $1/\tau_p$  increases by a factor  $\sim 30$  in proportion to  $T$  in the range 10 K to 100 K<sup>43</sup> due to acoustic phonon scattering<sup>66</sup>. The shortening of  $\tau_p$  towards 100 K thus results in a reduced DP dephasing rate with increasing temperature. A moderate increase of  $\tau_{KA}$  and constant behavior of  $\tau_{KB}$  when lowering the pump intensity is consistent with the rate contributions that we discussed here.

### C. Kerr signals from the 2DEG population

Figure 5 shows that population C with  $|g| \approx 0.34$  (besides a population with  $|g| \approx 0.44$ ) can be observed when the pump-photon density drops below  $\sim 10 \cdot 10^{11}$  photons/cm<sup>2</sup> per pulse. This crossover occurs around the 2DEG electron density due to doping, and this provides another indication that population C resides in the 2DEG quantum well. We assume again that the g-factor of this population is negative, and its value ( $\sim -0.34$ ) is then indeed consistent with the g-factor of electrons in heterojunction 2DEG systems and 15 to 20 nm wide GaAs QWs<sup>54,56,57</sup>. Other electrons popula-

tions that could be considered to give rise to the signal of population C can be convincingly rejected based on the g-factor values that we discussed before (Section III B). One can also rule out that population C corresponds to electrons or excitons trapped at impurities or defects in bulk GaAs, since a g-factor of this value has not been observed before by us or by others in bulk GaAs samples.

We further remark that the g-factor values  $|g| \approx 0.44$  and  $|g| \approx 0.34$  rule out the interpretation that observing two populations at these low pump intensities results from simultaneous probing of electrons in two different subbands of the heterojunction QW. Electrons in excited subbands have a g-factor that is closer to zero than the g-factor  $g \approx -0.34$  of electrons in the lowest subband<sup>61</sup>. Hence, the signal contribution with  $|g| \approx 0.44$  cannot result from 2DEG electrons. Such an interpretation would also be inconsistent with the observed decay times for the two populations in both TRKR traces and  $\Delta R$  traces. Instead, the population with  $|g| \approx 0.44$  must be due to electrons with bulk GaAs characteristics around the multilayer buffer, and we label it again with A. However, for these low pump intensities we were not able to determine whether this signal contribution is dominated by electrons just above (in layer II) or below (in layer I) the multilayer buffer (further discussed below).

We carried out two additional experiments which confirm that population C corresponds to 2DEG electrons. In the first experiment, we performed TRKR measurements on a heterostructure where the 2DEG had been removed by wet etching to a depth of about 120 nm. On such material, one can still observe populations A and B at high pump-photon densities. At low pump-photon densities, however, population C is no longer observed, and Kerr signals only contain a contribution with  $|g| \approx 0.44$ .

Secondly, we took TRKR data in an experiment where a microscope function<sup>62</sup> was included in the setup with Laser 1. In these experiments we measured with pump and probe spots with a diameter of  $1.6 \mu\text{m}$  that could be focussed at different (non-overlapping) locations on the sample. We measured on heterostructure material that had been processed into an ensemble of parallel wires of  $1.2 \mu\text{m}$  width at a periodicity of  $1.6 \mu\text{m}$ . The wires were realized with electron-beam lithography and subsequent wet etching to a depth ( $\sim 100 \text{ nm}$ ) that removes doping layer (and thereby the 2DEG) between wires. Figure 7 shows results that were obtained with this Kerr microscope. TRKR signals that were obtained with the pump and probe spot separated by  $4 \mu\text{m}$  distance but focussed on the same wire show both population A and C (beatings in Fig. 7b, and Fourier analysis gives g-factors 0.34 and 0.43). However, when repeating the measurement with the pump and probe separated by the same distance, but now separated along a direction orthogonal to the wires, the TRKR signals only show population A (no beatings in Fig. 7a, and Fourier analysis gives only  $|g| \approx 0.43$ ). This shows that population C must be in the accumulation layer, and that the signal contribution

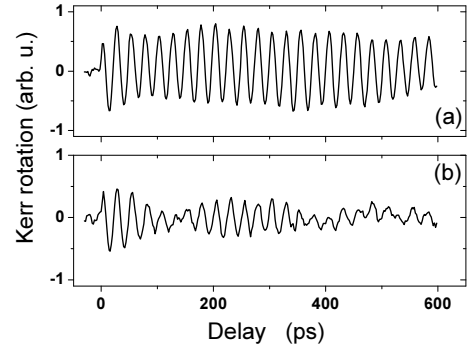


FIG. 7: TRKR signals obtained with a  $1.6 \mu\text{m}$  wide strip of 2DEG sample, obtained in a modified experimental setup where non-overlapping pump and probe spots of  $\sim 2 \mu\text{m}$  diameter were used. The trace in (a) is obtained with the pump focussed on the wire and the probe next to the wire with  $4 \mu\text{m}$  pump-probe separation. The trace in (b) is obtained with both the pump and probe focussed on the wire, again with  $4 \mu\text{m}$  pump-probe separation. Data obtained at 4.2 K and 7 Tesla, pump intensity of  $\sim 10 \cdot 10^{11}$  photons/ $\text{cm}^2$  per pulse.

with  $|g| \approx 0.43$  is (at least in the latter experiment) dominated by electrons in the substrate, just below the multilayer buffer. When pumping on a wire, photo-electrons in the accumulation layer (II) cannot escape from the wire: Escape into a deeper layer is blocked by the multilayer buffer, while lateral escape is blocked by a  $\sim 0.5 \text{ eV}$  barrier that results from upward bending of the conduction band below the etched trenches. Consequently, no population can appear in the accumulation layer (II) of neighboring wires. Photo-electrons below the multilayer buffer, however, can diffuse to areas below neighboring wires. Note that we cannot conclude from this experiment that population A is in the substrate (layer I) in our experiments with the larger and overlapping pump and probe spots as well. With the microscope we probe only those electrons that have migrated away from the pump area, and we pumped at a slightly higher intensity.

In the experiments with large overlapping spots, the clean segregation into two distinct signal components nevertheless points to the interpretation that population A resides in the substrate (and population C in the 2DEG). This interpretation then assumes that all photo-electrons that are induced throughout the accumulation layer (II) rapidly drift into heterojunction quantum well (see Fig. 1b), and the segregation between signal components A and C then results from the fact that the multilayer buffer blocks this drift for photo-electrons in the substrate. However, Salis *et al.* recently reported that the mobility of photo-electrons along the growth direction is surprisingly low at low pump-photon densities<sup>37</sup>. This means that photo-electrons near the multilayer buffer in the accumulation layer (II) do not migrate into the heterojunction quantum well within a few picoseconds. Instead, these electrons then have a dwell-time for staying in the deeper half of layer II that

is in excess of the observed Kerr signal decay times, and give a contribution to Kerr signals with  $|g| \approx 0.44$ . This scenario is also consistent with the fact that the amplitude of signal contribution A is comparable to that of C (unlike the results for the strong-pumping regime, where B gives a  $\sim 5$  times stronger contribution than A, see the discussion in Section III A). When population A is in the substrate (I), its signal is expected to be much weaker than the signal from C due to absorption throughout the accumulation layer and multiple reflections on the multilayer buffer (at least in the mono-color experiment, discussed below). This analysis point to the conclusion that at low pump-photon densities signal contribution A is dominated by photo-electrons just above the multilayer buffer (in layer II). However, we cannot fully rule a contribution from electrons below the multilayer buffer (in layer I), since the low mobility values are not yet fully understood.

For both the experiment with large laser spots and the microscope, we could again only observe two populations when pumping well above the bottom of the conduction band for bulk GaAs. With the pump wavelength longer than 808 nm, the Kerr signals were dominated by a single population with  $|g| \approx 0.44$ . The population with  $|g| \approx 0.34$  could be observed in both the two-color (Laser 1) and monocolour (Laser 2) experiment when pumping with 808 nm or shorter pump wavelength (see also the discussion in Section III B). However, different than for the experiments at high pump-photon densities, it is now not only a matter of having a short penetration depth for the pump. Obtaining Kerr signals with a strong contribution from 2DEG electrons probably also requires direct pumping into 2DEG subbands, and such transitions only obtain a significant matrix element when pumping at least  $\sim 20$  meV above the bottom of the conduction band for bulk GaAs. This clearly must play a role when the drift of photo-electrons into the heterojunction quantum well is indeed a surprisingly slow process at low pump intensities<sup>37</sup>.

Also for this low pump-photon density regime we studied how the Kerr signals depend on temperature  $T$  (presented in Fig. 6) and magnetic field  $B$  (only discussed). The Kerr signal decay times for populations A and C as a function of temperature are presented in Fig. 6c, and the corresponding g-factors in Fig. 6d, as obtained from fitting Eq. 3 on TRKR measurements taken at  $\sim 3.6 \cdot 10^{11}$  photons/cm<sup>2</sup> per pump pulse. The temperature dependence of the g-factor of population A follows again the trend that was reported for bulk GaAs<sup>38</sup>, and agrees with the high pump-photon density results when accounting for the lower photo-electron density. The trend for C shows a weaker temperature dependence, in agreement with a weaker dependence on  $E$  for 2DEG electrons (Eq. 1).

For the Kerr signal decay times (Fig. 6c), we found evidence that at 4.2 K the decay rate for both population A and C is dominated by the D'Yakonov-Perel' (DP) spin dephasing mechanism. We conclude this from fur-

ther experiments where our 2DEG material was etched into 1.2  $\mu\text{m}$  wide wires. The decay times then show a clear dependence on the crystal orientation of the wire. This proves that the DP mechanism dominates, with spin-orbit fields that are highly anisotropic in  $k$ -space. This results from a Rashba contribution<sup>2</sup> to the spin-orbit fields that can cancel the Dresselhaus contribution. The details of this study will appear in a future publication<sup>67</sup>. For the 2DEG population this conclusion agrees with earlier studies on high-mobility 2DEGs<sup>7,12,14</sup>. The fact that the decay time for C shows almost no dependence on temperature, indicates that the DP mechanism remains dominant up to 100 K, and that in this low pump-intensity regime the 2DEG mobility is not limited by scattering on acoustic phonons for all these temperatures. The Kerr signal decay times for population A are around 40 K longer than the electron-hole recombination times for bulk (as measured on bulk i-GaAs). This provides another indication that population A resides in a layer around the multilayer buffer where electron-hole recombination is suppressed due to band bending. The increase of the decay time for A when increasing the temperature up to 40 K indicates a suppression of the DP mechanism due to more rapid momentum scattering. Apparently, momentum scattering for this population is due to acoustic phonons. In the range 40 K to 100 K, the decay times shorten again. Here, the thermal spread in electron kinetic energies  $E$  becomes important, and results in a spread in g-factor values  $\Delta g$  that increases the precessional dephasing rate.

#### IV. SUMMARY AND CONCLUSIONS

We have characterized under what conditions it is possible to use time-resolved Kerr rotation measurements for studies of electron spin dynamics of 2DEG and epilayer ensembles in a GaAs/AlGaAs heterojunction system with a high-mobility 2DEG. Differences in g-factors of electrons in different layers of the heterostructure allow for discriminating the various populations, and for defining regimes where they behave as independent populations. This technique can be applied thanks to rapid momentum relaxation processes, which allows for pumping photo-electrons with an excess energy into the conduction band. At high pump-photon densities (above the 2DEG density from doping), this is crucial for having pump pulses with a short penetration depth, which ensures that Kerr detection is mainly probing electron populations near the wafer surface. At low pump-densities, where 2DEG spins can be observed, the excess pump-photon energy is also needed for directly pumping into subbands of the 2DEG. Furthermore, we find that having a barrier at about 1  $\mu\text{m}$  depth in the accumulation layer facilitates the segregation between Kerr signals. At high pump-photon densities this directly blocks the intermixing of accumulation-layer and substrate populations, and this drives the difference in g-factor. Here the accumu-

lation layer can be studied as a bulk epilayer filled with electrons. Further, since the Kerr response is enhanced by reflection on each interface in the heterostructure, the multilayer buffer may also be important at low pump-photon densities for realizing a segregation between the Kerr response from electrons in the heterojunction quantum well and electrons around the multilayer buffer.

We analyzed that in our experiments several mechanisms contribute to the dephasing of spin-oriented electron populations and the decay of Kerr signals, and complete understanding requires further studies. Our results here provide a basis for such further investigations, in particular for the spin dynamics of 2D electron ensembles in heterojunction quantum wells. An interesting direction is to explore whether confinement of such electron ensembles in micronscale device structures (as large quantum dots and wires) can be used to study spin dephasing anisotropy and suppression of spin dephasing due to confinement<sup>22,67</sup>. In addition, continuing our work with a microscope function in the setup allows for studying spin dynamics of 2DEG spins in transport channels with very high time resolution and micronscale spatial

resolution<sup>62</sup>. Our results thereby provide a path to performing studies on heterojunction 2DEG channels with tunable spin-orbit effects and electron density, which is interesting for work on the Datta-Das spin-transistor concept<sup>4</sup>. Notably, we showed that this can also be applied with non-overlapping pump and probe, giving access to exploring correlations between spin dynamics and spin transport.

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- <sup>50</sup> For the *n*-GaAs sample and bulk populations in the heterostructure sample, we observe *g*-factors close to  $|g| = 0.44$ . At the highest pump intensities, however, we observe values up to  $|g| \approx 0.42$ , as can be expected from band filling (see. Eq. 1). In addition, we observed for such high-pump-intensity traces that this effect diminishes over time scales of the electron-hole recombination times, resulting in weak *g*-factor drift during a TRKR trace that brings the *g*-factor back towards  $|g| \approx 0.44$ . We did not include this in our fitting as with Eq. 3, since it would give a much higher number of free parameters. However, this drift in-*g*-factor values only gave small shifts ( $< \pi$ ) in the phase of Kerr signals. We checked that the extracted  $\tau_K$  values with our approach were not significantly different from fitting results where these effects were included.
- <sup>51</sup> The decay time of the Kerr signal for *n*-GaAs is in this measurement substantially shorter than the long values observed in other studies<sup>5,32,33</sup>. This is due to the high pump intensity, and in exact agreement with the reported dependence on pump intensity<sup>5</sup>. With our setups we also observed Kerr signal decay times well in excess of 5 ns when using lower pump intensities and low fields.
- <sup>52</sup> We observe that using phases  $\phi_i$  as free parameters is needed for obtaining good fits. We also observed this with the *n*-GaAs reference material (but not when pumping at low intensities close to the bottom of the conduction band). This contradicts with the expected direction of spin orientation parallel to the pump beam at  $t = 0$ . However, we only use data from delays  $t > 5$  ps for fitting, since holes contribute to the Kerr signals up to  $t \approx 5$  ps. For  $t < 5$  ps there is also rapid relaxation of electron kinetic energy *E*. Since the effective *g*-factor has a value closer to zero for higher *E*, early energy relaxation is consistent with apparent phase offsets for  $t > 5$  ps. However, full justification of using  $\phi_i$  as free parameters requires further investigation.
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